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Superradiant Damping Effects in Photon-Echo Theory

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A theoretical model for superradiant damping effects in inhomogeneously broadened media and its application to photon-echo problems are discussed. Only optically thin systems are considered, and all nonradiative relaxation mechanisms are neglected. Numerical calculations are performed for a very wide square inhomogeneous line shape and square applied pulses. It is shown that excitation of the system becomes increasingly difficult as the superradiant lifetime τ_s becomes comparable with T_2^*. The energy reradiated after the end of the exciting pulse is considerable for $\tau_s \approx T_2^*$ and for some applied pulse areas. Two-pulse sequences produce multiple echoes as the microscopic polarizations spontaneously rephase. The first-echo amplitude maximum is found to occur for first-applied-pulse areas increasingly above $\frac{1}{2}\pi$ as τ_s decreases. For a given first-pulse area, the echo intensities increase initially as τ_s decreases (negligible damping) and finally decrease and vanish (important damping). The ratio of the second- to first-echo amplitudes is found to be proportional to T_2^*/τ_s for small superradiance levels and to decrease when τ_s approaches T_2^*. An approximate estimate of this ratio is derived for the region of linearity in T_2^*/τ_s.

I. INTRODUCTION

With the constant development of coherent optical-pulse techniques photon-echo experiments[1-4] will very likely become a valuable tool in the study of the evolution of the macroscopic polarization induced in a resonant medium by a light pulse. The direct interaction of the resonant atoms among themselves or with the host lattice may cause a loss of coherence and therefore the progressive destruction of the polarization. In this paper such nonradiative relaxation mechanisms are neglected and only cooperative radiation (superradiant[5]) damping effects are considered. The extent to which such damping effects take place depends on the degree of cooperation between the resonant atoms. The problem of superradiant damping effects on photon-echo intensities has been discussed elsewhere[6] on a very qualitative basis as no inhomogeneous broadening of the resonance line was considered. In the present paper our aim is to calculate the evolution of the macroscopic polarization induced by a two-pulse sequence in the presence of appreciable superradiant damping.

The theory[1,7] generally used to interpret the results of photon-echo experiments is based on two closely related fundamental assumptions. First, the resonant atoms do not "see" the field they emit themselves and are therefore coupled only to applied fields. Second, in the absence of an applied field the system does not (because of the first assumption) lose any energy but nevertheless radiates with intensity proportional to the square of the macroscopic polarization. For very large inhomogeneous broadening the macroscopic polarization is rapidly destroyed and the energy radiated is small. This removes any important contradiction in the second assumption. The approach just described will be called here the no-damping theory. It will not be applicable if the emission is so intense that most of the energy stored in the system is radiated, even in the short time before the destruction of the polarization. Clearly, a theory including radiation damping must be used when a large number of atoms cooperate in the spontaneous emission. Damping may be introduced by including in the Hamiltonian the interaction of the system with its own radiation field.[6,8,9] The formation of multiple echoes[1,3,10] after a two-pulse sequence can only be explained by the coupling of the system with the radiation emitted at the time of each echo. These comments show that superradiant damping effects must be considered in cases of strong echoes, particularly to calculate multiple echo intensities.

In the remainder of the paper the equations of motion are first derived so that inhomogeneously broadened resonance lines may be treated. Numerical results are discussed for very wide inhomogeneously broadened square line shapes and applied square pulses. The system is always supposed optically thin, so that the electric field may be considered uniform throughout the system. The response to a one-pulse excitation is calculated for different superradiant lifetimes τ_s as a preliminary to the photon-echo discussion. Two-pulse sequences are used to obtain multiple echoes whose intensities are analyzed as a function of first applied-pulse

area and τ_s. Finally, the results derived in this paper are discussed and compared with the available experimental data. In the Appendix the no-damping approach is used to calculate the intensity of the second echo.

II. EQUATIONS OF MOTION

In order to keep the problem as simple as possible we assume that the applied electric-field amplitudes are uniform over the entire system. The applied field is taken to be a plane wave propagating in the $\hat{z}$ direction and of the form

$$\vec{E}(z,t)=\hat{x}\,\mathcal{E}(t)\cos(\omega_0 t-kz)\ , \tag{1}$$

where $\hat{x}$ is the direction of the atomic electric transition dipole p. The system is composed of $\mathfrak{N}$ two-level atoms supposed identical but whose resonant frequencies ω have a distribution $g(\Delta\omega)$, where $\Delta\omega=\omega-\omega_0$. The frequency distribution is assumed symmetric and is defined such that

$$\int_{-\infty}^{+\infty} g(\Delta\omega)\,d\Delta\omega=1\ . \tag{2}$$

The applied field induces in each atom a polarization

$$\vec{P}(\Delta\omega,t)=\hat{x}p[u(\Delta\omega,t)\cos(\omega_0 t-kz) +v(\Delta\omega,t)\sin(\omega_0 t-kz)]\ , \tag{3}$$

where $u(\Delta\omega,t)$ and $v(\Delta\omega,t)$ are, respectively, the dispersion and the absorption components. We also define $w(\Delta\omega,t)$ as the population difference between the upper and lower levels which, multiplied by $\frac{1}{2}\hbar\omega$, gives the atomic-energy expectation value. We designate the average of $v(\Delta\omega,t)$ and $w(\Delta\omega,t)$ over all frequencies, respectively, by

$$\bar{v}(t)=\int_{-\infty}^{+\infty} g(\Delta\omega)v(\Delta\omega,t)\,d\Delta\omega\ , \tag{4}$$

$$\bar{w}(t)=\int_{-\infty}^{+\infty} g(\Delta\omega)w(\Delta\omega,t)\,d\Delta\omega\ . \tag{5}$$

Once excited, the system radiates spontaneously with intensity proportional to the square of the macroscopic polarization[5,6]

$$I(t)=(\mathfrak{N}\hbar\omega_0/2\tau_s)\,\bar{v}(t)^2\ . \tag{6}$$

The superradiant lifetime τ_s is inversely proportional to $\mathfrak{N}$ and is defined by

$$\tau_s^{-1}=(\mathfrak{N}/8\pi T_1)\int\left|e^{i(\vec{k}'-\vec{k})\cdot\vec{r}}\right|^2_{av}d\vec{k}'\ , \tag{7}$$

where $\vec{r}$ is the position vector of an atom within the system, and the average is taken over all atoms. In the no-damping approximation the macroscopic polarization is supposed to be destroyed by the dephasing of its individual components in a time so short that the energy radiated by the system according to Eq. (6) is negligible. The condition for this is that $T_2^*\ll\tau_s$. If this condition is violated the fraction of energy radiated becomes appreciable and a term leading to the proper radiation damping must be included in the equations of motion. This is done[6,8,9] by taking into account the interaction of the system with the field it spontaneously radiates, the amplitude of which is given by

$$\mathcal{E}_s(t)=-(\hbar/p\tau_s)\,\bar{v}(t)\ . \tag{8}$$

This field is added to any applied field present and the evolution of the system is described by the Bloch equations[9,11] in which the applied-field amplitude has been substituted by

$$\mathcal{E}^*(t)=\mathcal{E}(t)-(\hbar/p\tau_s)\,\bar{v}(t)\ . \tag{9}$$

This and the Bloch equations

$$\frac{du(\Delta\omega,t)}{dt}=-\Delta\omega v(\Delta\omega,t)\ , \tag{10}$$

$$\frac{dv(\Delta\omega,t)}{dt}=\Delta\omega u(\Delta\omega,t)-\frac{p}{\hbar}\,\mathcal{E}^*(t)w(\Delta\omega,t)\ , \tag{11}$$

$$\frac{dw(\Delta\omega,t)}{dt}=\frac{p}{\hbar}\,\mathcal{E}^*(t)v(\Delta\omega,t)\ , \tag{12}$$

are the equations of motion of the system. In the absence of an applied field the variation of the atomic energies is given by

$$\frac{dw(\Delta\omega,t)}{dt}=-\frac{1}{\tau_s}\,\bar{v}(t)v(\Delta\omega,t)\ . \tag{13}$$

It is then clear that, if $v(\Delta\omega,t)$ and $\bar{v}(t)$ have opposite signs, the energy of the atoms at $\Delta\omega$ *increases* during the spontaneous emission. However, the major contribution to the total energy comes from the atoms for which $v(\Delta\omega,t)$ and $\bar{v}(t)$ have the same sign, as can be seen by integrating Eq. (13) over all frequencies:

$$\tfrac{1}{2}\mathfrak{N}\hbar\omega_0\frac{d\bar{w}(t)}{dt}=-\frac{\mathfrak{N}\hbar\omega_0}{2\tau_s}\,\bar{v}(t)^2\ . \tag{14}$$

This, of course, is in agreement with the starting assumption of a spontaneous-emission intensity given by Eq. (6).

III. ONE-PULSE EXCITATION

In this section and in Sec. IV. we present and discuss the results of the numerical integration of the equations of motion. All applied pulses are square with duration τ_0 and variable area $\theta=(p/\hbar)\times\mathcal{E}\tau_0$. The inhomogeneous line shape is square and its width is always $T_2^{*-1}=80\tau_0^{-1}$. Time is given in units of τ_0. The superradiant lifetime is a parameter and may be approximately related to the resonant atom concentration N by the relation[1]

$$\tau_s\approx 8\pi T_1/\lambda^2 lN\ , \tag{15}$$

which, for a medium like ruby and for a sample thickness $l=0.01$ cm, gives

$$\tau_s\approx 10^9/N\ . \tag{16}$$

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In this section we discuss the response of the system to a one-pulse excitation, having always in mind the consequences to the response to a second pulse which is considered in Sec. IV. Before dealing with the case of important damping it is of some interest to use the no-damping limit to calculate which fraction of the energy stored in the system at the end of the applied pulse is subsequently radiated. For square applied pulses, the equations of motion with $\tau_s = \infty$ may be integrated exactly[12] and the fraction of energy radiated is easily calculated. The result is shown in Fig. 1 for the case of ruby, a concentration $N = 10^{18}$ atoms cm^{-3} and a sample thickness of 0.01 cm. For some applied-pulse areas, the effects of radiation loss may become considerable. At higher concentrations these effects are important even for small applied-pulse areas. In those circumstances the no-damping approximation is totally inadequate and the full equations of motion, (9) through (12), must be considered.

It has been shown[6] that, in the absence of inhomogeneous broadening and for important superradiant damping, the system is more difficulty excited by an applied pulse and subsequently decays to the ground state with emission of radiation. When inhomogeneous broadening is introduced we should expect it to prevent the damping in the limit of $\tau_s \gg T_2^*$ and to be of little effect in the opposite case. These two limits are therefore of no special interest,

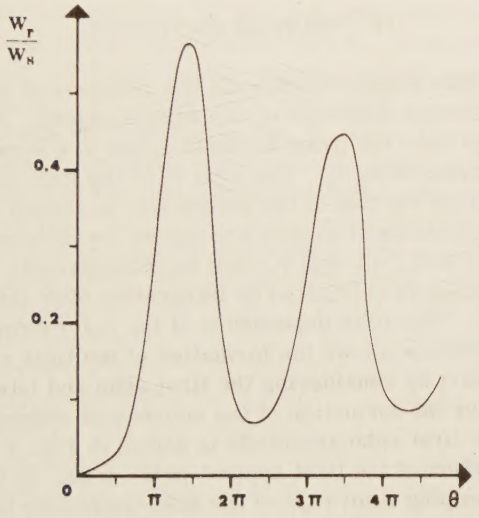

FIG. 1. Ratio of the energy radiated after the end of the applied pulse (W_r) to the energy stored in the system at the end of the pulse (W_s), plotted as a function of the applied pulse area. Parameters used: $N = 10^{18}$ atoms cm^{-3}, $l = 0.01$ cm, $T_1 = 4\times10^{-3}$ sec, $\lambda = 7\times10^{-5}$ cm, and $\tau_0 = 10^{-8}$ sec. As W_r is proportional to N^2 and W_s to N, the ratio is proportional to N and the results of the figure may be extended to other concentrations.

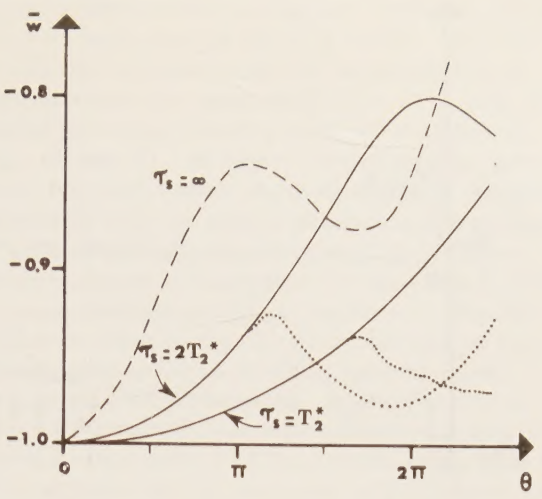

FIG. 2. Energy of the system, in units of $\frac{1}{2}\mathcal{N}\hbar\omega_0$, shown for different τ_s values as a function of the applied-pulse area. The full lines correspond to the energy at the end of the applied pulse and the dotted lines to the energy finally trapped in the system by the destruction of the macroscopic polarization. In the no-damping limit (dashed line) no energy is lost by the system after the end of the applied pulse.

and we will focus our attention on the region $\tau_s \approx T_2^*$. The equations of motion should be solved self-consistently but we have found that a one-iteration procedure is quite satisfactory for cases of $\tau_s \approx T_2^*$, in which the superradiant damping is not too strong. All results shown in the course of this paper for finite τ_s were obtained in this way. Figure 2 shows the average excitation energy at the end of the applied pulse (full line) and the average energy trapped in the system by the dephasing of the microscopic polarizations (dotted line), as a function of the applied-pulse area, for different values of τ_s. The dashed line represents the no-damping limit in which no energy is lost by the system. It is clear that the behavior of the system changes little until τ_s has decreased to $\approx T_2^*$. Once this order of magnitude has been attained a small decrease in τ_s leads to an appreciably larger difficulty in exciting the system and soon the excitation is appreciable only for large-area pulses.

It is of some interest to justify the thin-medium approximation used here. A simple calculation shows that, for the values of the parameters used in this paper, the absorbed fraction of the incident pulse energy is

$$\Delta W/W \approx (T_2^*/\tau_s)(\theta/\pi)^{-2}(\bar{w}+1) \ , \qquad (17)$$

where $\bar{w}$ is defined in Eq. (5). It is then clear (from Fig. 2) that the energy absorbed represents roughly 2–3% of the incident energy. The assump-

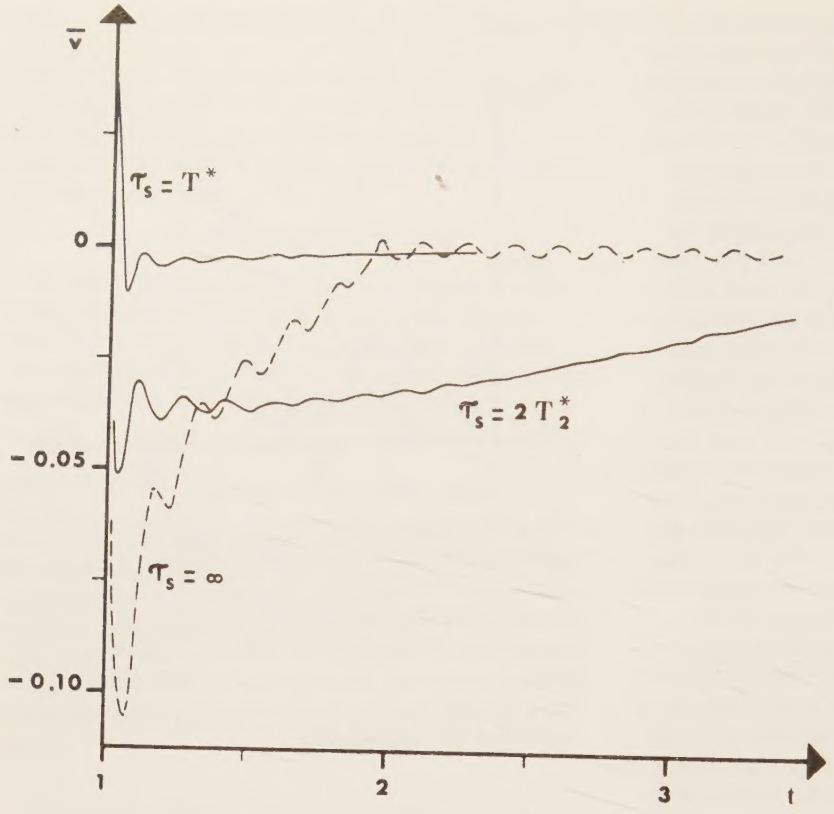

FIG. 3. Decay of the macroscopic polarization, in units of $\mathcal{N}p$, after the end of one applied 1.5π pulse. The dashed line represents the no-damping limit and has been reduced vertically by a factor of 2. Time is in units of τ_0. The period of the small oscillations is $4\pi T_2^*$.

tion of an optically thin medium is therefore seen to be a reasonable one.

The photon-echo amplitude measures, when there are no transverse relaxation mechanisms, the degree of excitation of the system at the time of the echo formation. Therefore we should expect the photon-echo dependence on the first-pulse area to be roughly similar to the dotted curves in Fig. 2. This is, of course, only a crude estimate, as the echo depends on the details of the reforming of the macroscopic polarization and not merely on the energy remaining in the system after the first pulse.

The macroscopic polarization decay is given in Fig. 3 for a 1.5π applied pulse and different τ_s values. The curve for the no-damping case is typical of very inhomogeneously broadened media.[13] The period of the oscillations is $\approx 4\pi T_2^*$, as seen in the figure. When T_2^* is decreased the oscillations disappear more rapidly[14] for $T_2^* = 0$. The origin of these oscillations is briefly discussed in the Appendix. With increasing superradiance the macroscopic polarization is initially smaller due to the smaller excitation achieved by the applied pulse but it can last much longer than in the no-damping limit.

IV. TWO-PULSE EXCITATION

In this section we discuss the response of the system to a sequence of two square pulses. Both pulses have the same duration τ_0 and are separated by a time delay τ_d. The area θ_1 of the first pulse is varied but that of the second one is always π. The equations of motion are solved for different values of θ_1, τ_d, and τ_s, and the macroscopic polarization is calculated by integration over the line shape. The time dependence of the macroscopic polarization shows the formation of multiple echoes. We start by considering the first echo and later discuss the formation of the subsequent echoes.

The first echo amplitude is shown in Fig. 4 as a function of the first applied-pulse area. In the no-damping limit ($\tau_s = \infty$) the echo-amplitude behavior is somewhat similar to the $\sin\theta_1$ dependence corresponding to the neglect of precession of the microscopic dipoles during excitation (Appendix). There is a linear behavior for small θ_1 and maxima near $\frac{1}{2}\pi$ and $\frac{3}{2}\pi$. The minimum near π is quite different from zero due to the precession of the atomic polarizations during the applied pulses. Also, the second maximum is more important than the first because, as is shown in the Appendix, the

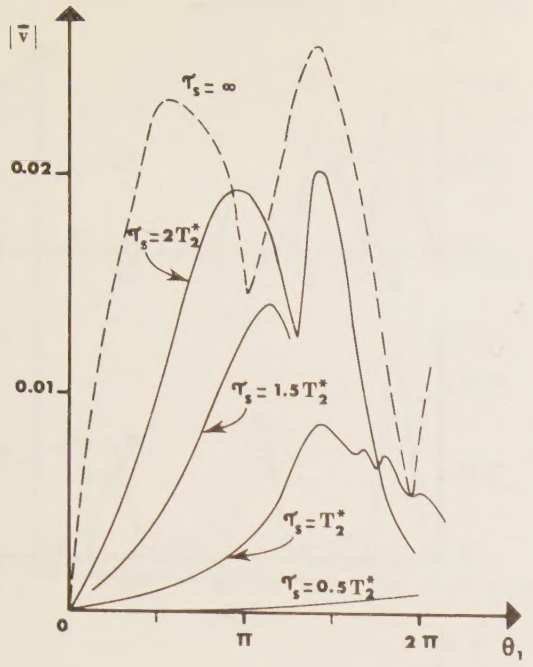

FIG. 4. Amplitude of the first photon echo, for different τ_s values, as a function of the first applied-pulse area. The dashed line represents the no-damping limit and has been reduced vertically by a factor of 2.

fraction of atoms excited by the first pulse increases with its area. If the superradiant lifetime is much larger than T_2^* the first echo amplitude does not change appreciably with τ_s. It is only when $\tau_s \approx T_2^*$ that sizable damping effects appear which are very sensitive to changes in τ_s. This trend can be observed in Fig. 4, where the introduction of important superradiance leads to the appearance of two distinctive features in the calculated curves. First, the linear region for small θ_1 shows a slope rapidly diminishing with τ_s and, as a related effect, the position of the first maximum is displaced towards larger θ_1 values. This is readily understood in terms of the increasing difficulty of the pulse in exciting the system and of the larger ability of the system to reradiate spontaneously. The echo-amplitude maximum corresponds approximately to the maximum energy trapped in the system after the first pulse (Fig. 2). Second, the curves for $\tau_s \approx T_2^*$ in Fig. 4 show an inflection point after the linear region, which is a characteristic of important radiation-damping cases. It should be noted that this behavior is independent of any absolute-intensity considerations either of the applied pulses or of the echo.

We consider now the question of the echo-intensity dependence on the number of cooperating resonant atoms. The geometry of the system and the applied-pulse areas are supposed constant, but the resonant atom concentration is varied. At sufficiently low concentrations the superradiance is negligible and $\bar{v}$ is independent of N. The echo intensity increases therefore with N^2 according to Eqs. (6) and (7). At higher concentrations, however, the superradiant damping begins to compete effectively with the applied pulses and $\bar{v}$ decreases. At sufficiently high concentrations the N^2 dependence no longer compensates for the lower $\bar{v}$, and the echo intensities begin to decrease. This behavior is shown in Fig. 5, where the dashed line corresponds to the no-damping approximation. For larger first pulse areas, smaller values of τ_s are necessary for the superradiant damping to become comparable with the induced excitation. This explains why the maximum echo intensity appears at increasingly higher concentrations when θ_1 increases.

We have seen in Sec. III that the spontaneous emission following the applied pulse may last as long as $\approx 10^2 \times T_2^*$ (Fig. 3). As a consequence we have found a dependence of the first echo amplitude on the applied-pulse separation for $\tau_d \lesssim 3\tau_0$. For larger values of τ_d the echo remains unchanged when τ_d is varied, as the spontaneous emission following the first applied pulse has ceased when the second pulse is applied.

In the remainder of this discussion we are concerned with the problem of multiple echo formation. In the no-damping limit, if only one pair of

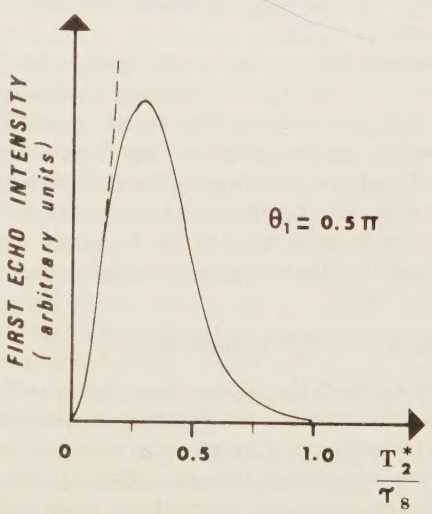

FIG. 5. First-echo intensity in arbitrary units, for $\theta_1 = 0.5\pi$, as a function of T_2^*/τ_s. For a ruby sample 0.01 cm thick $N \approx 10^{19} \times T_2^*/\tau_s$. For low superradiant damping the curve is approximately a parabola. The dashed line was calculated with the no-damping assumption.

pulses is applied to the system, the macroscopic polarization will reappear to form only one echo (Appendix). The experimental observation of multiple echoes[1, 3, 10] leads therefore to the conclusion that the interaction of the system with the first echo radiation must be responsible for the formation of subsequent echoes. But the interaction of the system with its own radiation is precisely the mechanism of radiation damping. The no-damping approximation can be used to give the first echo more or less accurately, but the multiple echo formation appears naturally in the evolution in time of the macroscopic polarization only with the superradiant damping approach.

Examples of calculated multiple echoes are shown in Fig. 6, where the time origin is chosen such that the first echoes coincide in both cases. They have been calculated by solving the equations of motion when two pulses are applied. The separation between echoes is $\tau_d + \frac{1}{2}\tau_0$, which accounts for the delay of the third echo for $\tau_d = 3\tau_0$ with respect to the second echo for $\tau_d = 6\tau_0$. The $\bar{v}(t)$ values for successive echoes are of opposite signs for θ_1 corresponding to the second lobe of the curves in Fig. 4 and of the same sign for smaller θ_1 values. In general each echo is accompanied by considerable ringing[15] as in Fig. 6(b) and this is the cause of the slight deformations of the echoes in Fig. 6(a). The lower part of that figure shows the energy loss accompanying each echo. The ratio of second to first echo amplitudes is shown in Fig. 7 as a function of T_2^*/τ_s for $\theta_1 = 0.5\pi$ and $\tau_d = 3\tau_0$. At low superradiance levels the variation is linear in T_2^*/τ_s.

Multiple echoes can be obtained in the linear region if we use the no-damping approach and neglect the precession of the atomic polarizations during the applied pulses. The first echo can be considered as a third applied pulse and the response of the system to the three-pulse sequence calculated analytically as is done in the Appendix. The ratio of second to first echo amplitudes is then found to be [Eq. (A9)]

$$v_{2e}/v_{1e} \approx \tfrac{1}{2}(T_2^*/\tau_s)\sin\theta_1 \sin\theta_2 \ , \qquad (18)$$

where we have the linear dependence in T_2^*/τ_s. From this equation the relative signs of the echo polarizations can also be determined in accordance with our numerical results. This is, of course, only an approximate calculation, even in the limit of small superradiance, as the important dephasing of the atomic polarizations is neglected during the applied pulses. At high superradiance levels the result of Eq. (18) is not applicable and we find that the ratio goes through a maximum and then decreases. This is explained by the increasingly important radiation damping during the formation

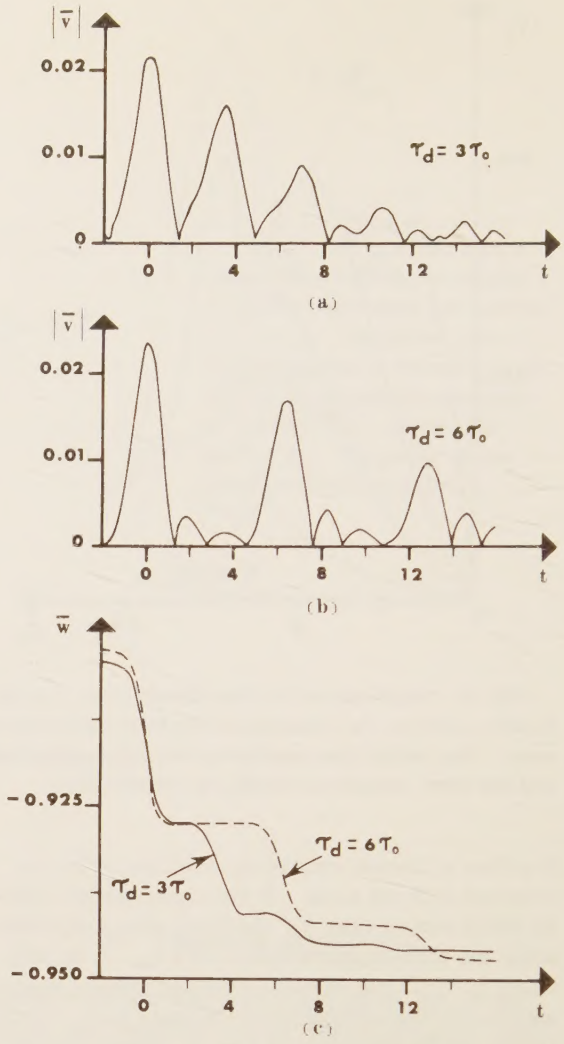

FIG. 6. Multiple echoes for $\tau_s = 2T_2^*$ and $\theta_1 = 1.5\pi$, as a function of time. Time is in units of τ_0 and the origin was chosen at $2(\tau_0 + \tau_d)$ to show the first echoes coincident. The multiple echo amplitudes are shown in (a) for $\tau_d = 3\tau_0$ and in (b) for $\tau_d = 6\tau_0$. The separation between echoes is $\tau_d + \frac{1}{2}\tau_0$. The energy of the system, in units of $\frac{1}{2}\mathfrak{N}\hbar\omega_0$ is shown in (c).

of each echo. At high superradiance levels the system is considerably less excited at the time of the second echo and will radiate correspondingly less.

V. DISCUSSION

We have seen that the no-damping approximation is not adequate above a certain concentration of resonant atoms because of the appearance of important cooperative effects. The Bloch equations, including a damping term, were used to calculate photon echo intensities, in particular in the region

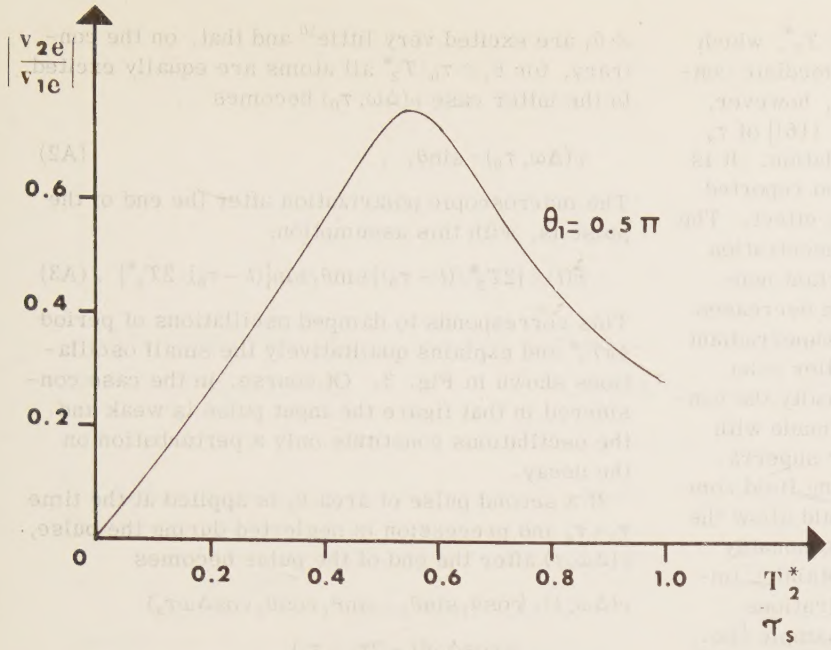

FIG. 7. Ratio of second to first photon-echo amplitudes for $\theta_1 = 0.5\pi$, as a function of T_2^*/τ_s. For a ruby sample 0.01 cm thick $N \approx 10^{19} \times T_2^*/\tau_s$. For low superradiant damping the ratio increases linearly.

$\tau_s \approx T_2^*$. The main results of the present paper may be summarized as follows: (a) The dependence of the first echo intensity on the first applied-pulse area was calculated for different τ_s values. The first maximum of echo intensity corresponds to first pulse areas increasingly above $\frac{1}{2}\pi$ as τ_s decreases. (b) For a given first pulse area, the echo intensity increases initially with N^2, goes through a maximum, and vanishes at higher concentrations. (c) The model used in the calculations leads to the formation of multiple echoes as the macroscopic polarization spontaneously evolves after two applied pulses. The ratio of second to first echo amplitudes first increases linearly with concentration, goes through a maximum and decreases at higher concentrations. This is in agreement with a model for the formation of the second echo in the small superradiant damping limit, in which the two applied pulses and the first echo act as a sequence of three equally spaced pulses.

The calculations presented in this paper were aimed at predicting the results of experiments performed in conditions similar to those described in Ref. 1 for the case of ruby. The experiments reported to date in the literature deal mainly with the study of nonradiative relaxation mechanisms. We discuss below the available experimental data relevant to the present study. These are unfortunately very few, but it seems to us that more data could be obtained from experiments similar to those already reported. More specifically, a systematic study of echo intensities as a function of exciting pulse intensity and concentration would be needed.

The first result of this paper which we discuss is the echo intensity dependence on the first pulse area. Compaan, *et al.*[4] found that the echo intensity was linear in the first pulse intensity at low intensities but that it increased more slowly at higher intensities. This behavior does not correspond to a superradiance high enough to introduce an inflection point in the intensity curve, but could correspond nevertheless to τ_s of the order of $1.5 \times T_2^*$ or $2T_2^*$. The other clue to the presence of superradiance damping would be the position of the intensity maximum. To obtain this, the area of first pulse must be calibrated in a low concentration experiment (where the echo intensity maximum appears for $\theta_1 \approx 0.5\pi$). This information will then allow to determine whether, at higher concentrations, the maximum occurs for $\theta_1 > 0.5\pi$. It is also possible that experiments at higher concentrations than that used in Ref. 4 ($N \approx 10^{19}$ atoms cm^{-3}) would show the inflection point characteristic of strong superradiant damping.

The second point to be considered is the first echo-intensity dependence on concentration for a given value of θ_1. Experiments of this type are more reliable with gaseous media (where the pressure of the active gas can be easily changed), but have also been performed in ruby.[1,16] It is reported that, when the concentration is increased 10 and 100 times from $N \approx 10^{18}$, the echo increases but appears to "saturate" for the highest concentration.[16] It seems that the first pulse area used[17] was approximately 2π. In that case our calcula-

tions show the echo maximum near $\tau_s = T_2^*$, which should correspond roughly to the intermediate concentration mentioned above. It should, however, be remembered that our estimate [Eq. (16)] of τ_s is merely an order of magnitude calculation. It is possible that the saturation[18] of the echo reported corresponds to a superradiant damping effect. The difficulty in interpreting the higher concentration results comes from the fact that important nonradiative relaxation takes place[19] which decreases the echo intensity. This, rather than superradiant effects, could be the origin of the smaller echo intensity increase. To avoid this difficulty the concentration dependence study should be made with smaller θ_1 values. In that case, lower superradiance levels are needed to give damping field comparable with the applied one. This would allow the detection of superradiant damping at reasonably low concentrations. Another way of obtaining important superradiance at lower concentrations would be to increase the length of the sample [Eq. (15)]. In any case, the nonradiative relaxation effects can always be subtracted by changing τ_d and extrapolating the results to $\tau_d = 0$ (as in Ref. 19).

Multiple echoes have been observed in ruby.[1] The second echo shown in Ref. 1 is approximately half as intense as the first one, showing the presence of appreciable superradiance. However, no information is given on the variation of multiple echo intensities with concentration. This type of information would allow the determination of τ_s, which could then be compared with the values obtained from the first echo behavior.

From the above discussion of the published experimental results we may conclude that no evidence of important superradiance (other than the detection of the echoes themselves) exists. On the other hand, it is clear that none of the experiments reported contradicts the calculations presented in this paper. We have proposed experiments to check our theoretical model, which appear to present no more difficulty than those already performed.

APPENDIX

The resolution of the Bloch equations in the no-damping limit can be done analytically for the case of square applied pulses.[12] The comportment of the system in these conditions is well known, but a few remarks are useful in understanding the much more complicated situation of important radiation damping. If a first pulse of area θ_1 is applied, $v(\Delta\omega, t)$ at the end of the pulse is given by

$$v(\Delta\omega, \tau_0) = \theta_1[(\Delta\omega\tau_0)^2 + \theta_1^2]^{-1/2} \times \sin[(\Delta\omega\tau_0)^2 + \theta_1^2]^{1/2} \quad . \tag{A1}$$

It is easily seen that those atoms for which $\Delta\omega\tau_0 \gg \theta_1$ are excited very little[20] and that, on the contrary, for $\theta_1 \gg \tau_0/T_2^*$ all atoms are equally excited. In the latter case $v(\Delta\omega, \tau_0)$ becomes

$$v(\Delta\omega, \tau_0) = \sin\theta_1 \quad . \tag{A2}$$

The macroscopic polarization after the end of the pulse is, with this assumption,

$$\bar{v}(t) = [2T_2^*/(t - \tau_0)] \sin\theta_1 \sin[(t - \tau_0)/2T_2^*] \quad . \tag{A3}$$

This corresponds to damped oscillations of period $4\pi T_2^*$ and explains qualitatively the small oscillations shown in Fig. 3. Of course, in the case considered in that figure the input pulse is weak and the oscillations constitute only a perturbation on the decay.

If a second pulse of area θ_2 is applied at the time $\tau_0 + \tau_d$ and precession is neglected during the pulse, $v(\Delta\omega, t)$ after the end of the pulse becomes

$$v(\Delta\omega, t) = (\cos\theta_1 \sin\theta_2 + \sin\theta_1 \cos\theta_2 \cos\Delta\omega\tau_d) \times \cos\Delta\omega(t - 2\tau_0 - \tau_d) - \sin\theta_1 \sin\Delta\omega\tau_d \sin\Delta\omega(t - 2\tau_0 - \tau_d) \quad . \tag{A4}$$

This result may be written, neglecting free damping terms,

$$v(\Delta\omega, t) = \sin\theta_1(\cos\theta_2 - 1) \times \cos\Delta\omega(t - 2\tau_0 - 2\tau_d) \quad . \tag{A5}$$

An echo is formed at a time $2(\tau_0 + \tau_d)$ with the familiar amplitude dependence on the applied-pulse areas.[1,7] In the no-damping theory this is the only echo which is formed. If, however, we make one step in the direction of the inclusion of radiation damping by considering the first echo as an applied pulse, further echoes may be obtained. Let then a third pulse (the first echo) of area θ_{1e} and duration τ_0 be applied at a time $2\tau_d + \frac{3}{2}\tau_0$, so that its center coincides with the first echo maximum. Each $v(\Delta\omega, t)$ after the end of this pulse is, neglecting free decay terms,

$$v(\Delta\omega, t) = -\sin\theta_2(\tfrac{1}{2}\sin\theta_1 \sin\theta_{1e} + \sin^2\tfrac{1}{2}\theta_{1e} \cos\theta_1) \cos\Delta\omega(t - 3\tau_d - \tfrac{5}{2}\tau_0) - \sin\theta_1 \cos^2\tfrac{1}{2}\theta_2 \sin^2\tfrac{1}{2}\theta_{1e} \times \cos\Delta\omega(t - 4\tau_d - \tfrac{5}{2}\tau_0) \quad . \tag{A6}$$

We see that two additional echoes are formed at times $3\tau_d + \frac{5}{2}\tau_0$ and $4\tau_d + \frac{5}{2}\tau_0$. As the first echo intensity is in general small, we can substitute the $\sin\theta_{1e}$ and $\sin\frac{1}{2}\theta_{1e}$ by their arguments and may neglect the two last terms if neither $\sin\theta_1$ nor $\sin\theta_2$ are very small. Only the second echo is then important and its amplitude[21] is given by

$$v_{2e} = -\tfrac{1}{2}\theta_{1e} \sin\theta_1 \sin\theta_2 \quad . \tag{A7}$$

According to Eq. (8) the first echo area is

$$\theta_{1e} \approx -(T_2^{*}/\tau_0)(\tau_0/\tau_s)v_{1e} \ . \qquad \text{(A8)}$$

which leads to

$$v_{2e}/v_{1e} \approx \tfrac{1}{2}(T_2^{*}/\tau_s)\sin\theta_1 \sin\theta_2 \ . \qquad \text{(A9)}$$

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[15] Calculations performed in the no-damping approximation show that the ringing is not a superradiant effect. The oscillations in the macroscopic polarization are a consequence of the inhomogeneous broadening, which prevents the rephasing of all the dipoles at the same time. For some first pulse areas the ringing does not appear, both with and without superradiant damping effects.

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[18] We have seen that, for the dependence of τ_s on the concentration used in this paper [Eq. (16)], the attenuation of the applied pulses was small and roughly independent of N. However, if the τ_s values encountered in an actual experiment are larger than those given by Eq. (16) the pulse attenuation is greater. In any case, the attenuation of the pulses will be roughly independent of the concentration: As the concentration increases more atoms absorb energy, but each one absorbs less because of the increase in superradiant damping. We may then conclude that, even though appreciable pulse attenuation may take place, this does not cause the saturation of the echo at higher concentrations. The argument presented here is clearly only valid in the range $\tau_s \approx T_2^{*}$ discussed in this paper. In the no-damping limit the attenuation of the pulses increases with N.

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[20] The fraction of atoms appreciably excited by the pulse is then $f \approx \Delta\omega T_2^{*} \approx \theta_1 T_2^{*}/\tau_0$. For very intense pulses of the same duration all atoms will be equally excited (f 1) and for low intensity pulses $f < T_2^{*}/\tau_0$. It is only for $\theta_1 \approx 1$ that the familiar value $f \approx T_2^{*}/\tau_0$ is found.

[21] The formation of the second echo is sometimes (see, for example, Ref. 1) attributed to the action of the second applied pulse and of the first echo as a two-pulse sequence leading to another echo. It can be seen that such a mechanism leads only to the second term in Eq. (A4) which is much smaller than the first one because of its θ_{1e}^2 dependence. Also, such an explanation gives a ratio of first to second echo amplitudes proportional to the square of T_2^{*}/τ_s, in contradiction with the results of the exact calculation.